

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In the application of

STEVEN LUO

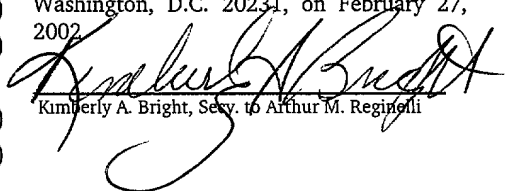
Serial No.

Filed

For PREPARATION OF CONJUGATED DIENE)  
POLYMERS BY USING AN IRON-BASED )  
CATALYST SYSTEM )

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Assistant Commissioner for Patents,  
Washington, D.C. 20231, on February 27,  
2002.

  
Kimberly A. Bright, Secy. to Arthur M. Reginaldi

**PRELIMINARY AMENDMENT**

ASSISTANT COMMISSIONER FOR PATENTS

Washington, D.C. 20231

Sir:

**In the specification:**

At page one, after the title, please insert the following.

This application is a continuation of U.S. Serial No. 09/475,343,  
filed on December 30, 1999, which is a continuation-in-part of U.S. Serial Nos.  
09/172,305, now U.S. Patent No. 6,277,779, 09/173,956, now U.S. Patent No.  
6,180,734, and 09/439,861, now U.S. Patent No. 6,211,313.

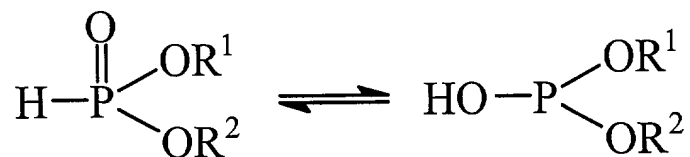
At page 16, lines 13-20, please replace the following paragraph. A  
marked up paragraph has also been attached.

In another preferred embodiment of the present invention where it  
is especially desirable to synthesize syndiotactic 1,2-polybutadiene, the molar ratio  
of the organoaluminum compound to the iron-containing compound (Al/Fe)  
should be relatively high. For purposes of this specification, the term "relatively  
high" generally refers to an Al/Fe molar ratio that can be varied from about 10:1  
to about 100:1, more preferably from about 13:1 to about 40:1, and even more  
preferably from about 14:1 to about 30:1, with it being understood that this ratio  
can vary as described hereinbelow.

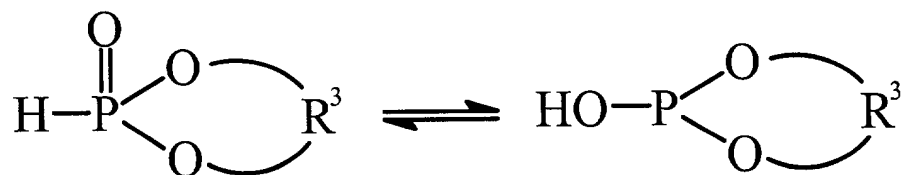
**In the claims:**

Please cancel claims 1-20 without prejudice or disclaimer, but before doing so, please add the following claims.

21. A process for preparing conjugated diene polymers comprising the step of:  
polymerizing monomer consisting essentially of conjugated diene monomer in the presence of a catalyst composition that is formed by combining:
- (a) an iron-containing compound;
  - (b) a hydrogen phosphite; and
  - (c) an organoaluminum compound, where the catalyst composition includes from about 0.01 to 1.0 mmol of the iron-containing compound per 100 grams of monomer, and where said step of combining occurs in the presence of at least one type of conjugated diene monomer.
22. The process of claim 21, where the molar ratio of the organoaluminum compound to the iron-containing compound (Al/Fe) is from about 1:1 to about 100:1.
23. The process of claim 22, where the molar ratio of the hydrogen phosphite to the iron-containing compound (P/Fe) is from about 0.5:1 to about 50:1.
24. The process of claim 21, where the conjugated diene monomer includes 1,3-butadiene monomer.
25. The process of claim 24, where the molar ratio of the organoaluminum compound to the iron-containing compound (Al/Fe) is from about 1:1 to about 100:1.
26. The process of claim 21, where the hydrogen phosphite is an acyclic hydrogen phosphite defined by the following keto-enol tautomeric structures:

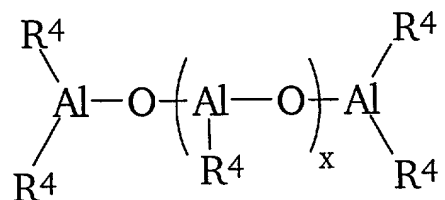


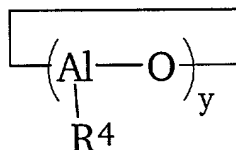
or a cyclic hydrogen phosphite defined by the following keto-enol tautomeric structures:



or a mixture thereof, where  $\text{R}^1$  and  $\text{R}^2$ , which may be the same or different, are mono-valent organic groups, and where  $\text{R}^3$  is a divalent organic group.

27. The process of claim 21, where the organoaluminum compound comprises at least one compound defined by the formula  $\text{AlR}_n\text{X}_{3-n}$ , where each R, which may be the same or different, is a mono-valent organic group, where each X, which may be the same or different, is a hydrogen atom, a carboxylate group, an alkoxide group, or an aryloxy group, and where n is an integer including 1, 2 or 3, or where the organoaluminum compound comprises at least one compound defined by one of the following formulas:





where x is an integer of 1 to about 100, y is an integer of 2 to about 100, and each R<sup>4</sup>, which may be the same or different, is a mono-valent organic group.

28. The process of claim 21, where the organoaluminum compound comprises trihydrocarbylaluminum, dihydrocarbylaluminum hydride, hydrocarbylaluminum dihydride, dihydrocarbylaluminum carboxylate, hydrocarbylaluminum bis(carboxylate), dihydrocarbylaluminum alkoxide, hydrocarbylaluminum dialkoxide, dihydrocarbylaluminum aryloxide, hydrocarbylaluminum diaryloxide, or mixtures thereof.

29. The process of claim 21, where the catalyst composition is formed by first combining the iron-containing compound and the hydrogen phosphite in the presence of the at least one type of conjugated diene monomer to form an initial composition, followed by combining the initial composition with the organoaluminum compound and, optionally, additional conjugated diene monomer.

30. The process of claim 21, where the catalyst composition is formed by first combining the iron-containing compound and the organoaluminum compound in the presence of the at least one type of conjugated diene monomer to form an initial composition, followed by combining the initial composition with the hydrogen phosphite and, optionally, additional conjugated diene monomer.

31. The process of claim 21, where the catalyst composition is formed by first combining the iron-containing compound and the organoaluminum compound outside the presence of the at least one type of conjugated diene monomer to form an initial composition, followed by combining the initial composition with the

hydrogen phosphite in the presence of the at least one type of conjugated diene monomer.

32. The process of claim 21, where the catalyst composition is formed by first combining the iron-containing compound and the hydrogen phosphite outside the presence of the at least one type of conjugated diene monomer to form an initial composition, followed by combining the initial composition with the organoaluminum compound in the presence of the at least one type of conjugated diene monomer.

33. The process of claim 21, where the catalyst composition includes from about 0.02 to about 0.5 mmol of the iron-containing compound per 100 grams of monomer.

34. The process of claim 33, where the catalyst composition includes from about 0.05 to about 0.5 mmol of the iron-containing compound per 100 grams of monomer.

35. The process of claim 21, where the conjugated diene monomer consists of 1,3-butadiene monomer.

36. The process of claim 21, where the catalyst composition is formed by first combining the hydrogen phosphite and the organoaluminum compound in the presence of the at least one type of conjugated diene monomer to form an initial composition, followed by combining the initial composition with the iron-containing compound and, optionally, additional conjugated diene monomer.

37. The process of claim 21, where the catalyst composition is formed by first combining the hydrogen phosphite and the organoaluminum compound outside the presence of the at least one type of conjugated diene monomer to form an initial composition, followed by combining the initial composition with the iron-

containing compound in the presence of the at least one type of conjugated diene monomer.

38. A process for preparing syndiotactic 1,2-polybutadiene, the process comprising:

polymerizing monomer consisting essentially of 1,3-butadiene monomer in the presence of a catalyst composition that is formed by combining:

- (a) an iron-containing compound;
- (b) a hydrogen phosphite; and
- (c) an organoaluminum compound, where the molar ratio of the organoaluminum compound to the iron-containing compound (Al/Fe) is 12:1 or greater, and where said step of combining occurs in the presence of at least one type of conjugated diene monomer.

39. The process of claim 38, where the molar ratio of the organoaluminum compound to the iron-containing compound (Al/Fe) is 13:1 to about 40:1.

40. The process of claim 39, where the molar ratio of the organoaluminum compound to the iron-containing compound (Al/Fe) is 14:1 to about 30:1.

41. The process of claim 38, where the catalyst composition is formed by first combining the iron-containing compound and the hydrogen phosphite in the presence of the at least one type of conjugated diene monomer to form an initial composition, followed by combining the initial composition with the organoaluminum compound and, optionally, additional conjugated diene monomer.

42. The process of claim 38, where the catalyst composition is formed by first combining the iron-containing compound and the organoaluminum compound in the presence of the at least one type of conjugated diene monomer to form an initial composition, followed by combining the initial composition with the

hydrogen phosphite and, optionally, additional conjugated diene monomer.

43. The process of claim 38, where the catalyst composition is formed by first combining the iron-containing compound and the organoaluminum compound outside the presence of the at least one type of conjugated diene monomer to form an initial composition, followed by combining the initial composition with the hydrogen phosphite in the presence of the at least one type of conjugated diene monomer.

44. The process of claim 38, where the catalyst composition is formed by first combining the iron-containing compound and the hydrogen phosphite outside the presence of the at least one type of conjugated diene monomer to form an initial composition, followed by combining the initial composition with the organoaluminum compound in the presence of the at least one type of conjugated diene monomer.

45. The process of claim 38, where the catalyst composition is formed by first combining the hydrogen phosphite and the organoaluminum compound in the presence of the at least one type of conjugated diene monomer to form an initial composition, followed by combining the initial composition with the iron-containing compound and, optionally, additional conjugated diene monomer.

46. The process of claim 38, where the catalyst composition is formed by first combining the hydrogen phosphite and the organoaluminum compound outside the presence of the at least one type of conjugated diene monomer to form an initial composition, followed by combining the initial composition with the iron-containing compound in the presence of the at least one type of conjugated diene monomer.

#### REMARKS

No fee is believed due with the filing of this document, however, in the event that a fee required for the filing of this document is missing or insufficient, the undersigned attorney hereby authorizes the Commissioner to

charge payment of any fees associated with this communication or to credit any overpayment to Deposit Account No. 06-0925.

Respectfully submitted,

  
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February 27, 2002



**MARKED-UP PARAGRAPH**

In another preferred embodiment of the present invention where it is especially desirable to synthesize syndiotactic 1,2-polybutadiene, the molar ratio of the organoaluminum compound to the iron-containing compound (Al/Fe) should be relatively high. For purposes of this specification, the term "relatively high" generally refers to an Al/Fe molar ratio that can be varied from about 10:1 to about 100:1, more preferably from about [13.1] 13:1 to about 40:1, and even more preferably from about 14:1 to about 30:1, with it being understood that this ratio can vary as described hereinbelow.